

A new model to describe the physics of $(\text{VO})_2\text{P}_2\text{O}_7$

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In the past different models for the magnetic salt vanadyl pyrophosphate $(\text{VO})_2\text{P}_2\text{O}_7$ were discussed. Neither a spin ladder nor an alternating chain are capable to describe recently measured magnetic excitations. In this paper we propose a 2D model that fits better to experimental observations.

PACS numbers: 75.10.-b, 75.25.+z, 75.40.Mg

Low dimensional quantum spin systems have been a field of intense theoretical and experimental research over the last decades. Special interest was given to spin ladder and chain materials. One compound that has been examined in this context is the insulating magnetic salt vanadyl pyrophosphate $(\text{VO})_2\text{P}_2\text{O}_7$. Initially it was considered as a prototypical realization of a two-leg antiferromagnetic Heisenberg ladder [1]. However, susceptibility data on polycrystalline and single crystalline material could be well fitted with both, ladder or alternating chain models [1–3], stressing the fact that susceptibility is not too sensitive to the particular model. Early inelastic neutron scattering measurements on polycrystalline samples indicated a spin gap of about 3.7 meV and supported a two-leg ladder model with the coupling constants estimated from susceptibility data [4].

Recent neutron scattering experiments with powder samples [5] and with an array of single crystals [6] provided detailed information on the low-energy excitation spectrum. Garrett et al. [6] observed a triplet branch with strongest (antiferromagnetic) dispersion in b-direction, weak (ferromagnetic) dispersion in a-direction, and a spin gap of 3.1 meV. Most notably they found an additional *second* branch, separated from the first by an energy smaller than the gap. This was inconsistent with the picture, of $(\text{VO})_2\text{P}_2\text{O}_7$ being a spin ladder in a-direction, but also an alternating Heisenberg chain in b-direction can not explain a second triplet branch over the whole Brillouin zone, as was shown recently [7,8].

In this work, starting with the alternating Heisenberg chain, we check whether coupling of (two) chains resolves this puzzling situation. As we do not succeed proceeding this way, we consider a new, truly two-dimensional model. We perform exact diagonalizations of finite systems with up to 32 spins and periodic boundary conditions, supplemented by finite-size analysis if possible.

The Hamiltonian of the alternating Heisenberg chain (AHC) reads as follows

$$H_{\text{AHC}} = J_b \sum_i (1 + \delta(-1)^i) \mathbf{S}_i \cdot \mathbf{S}_{i+1}, \quad (1)$$

where \mathbf{S}_i are spin- $\frac{1}{2}$ operators and i denotes the sites in b-direction (see Fig. 1). For $\delta > 0$ the spectrum has a gap; there is an one-magnon branch and a singlet branch, at

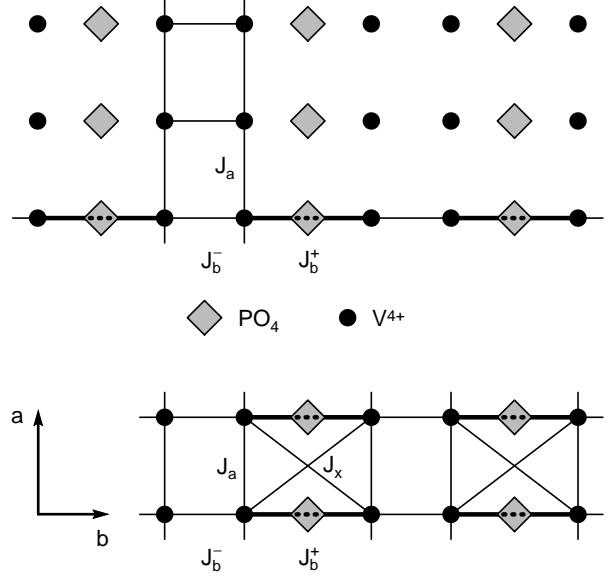


FIG. 1.: Schematic structure of $(\text{VO})_2\text{P}_2\text{O}_7$. The exchange couplings are depicted for (i) the ladder model ($J_{\parallel} = J_a, J_{\perp} = J_b$), (ii) the alternating chain model ($J_b^\pm = J_b(1 \pm \delta)$), and (iii) the new model (J_b^\pm, J_a, J_x). Throughout we measure energies in units of J_b .

least around momentum $\frac{\pi}{2}$, below a continuum of states.

As an example Fig. 2 shows the low-lying excitations of a finite system of 32 sites for $\delta = 0.2$. The magnon branch is fitted to a sum of cosines $\omega_q^m = \sum_{n=0}^5 a_n \cos(2nq)$ and the shaded region corresponds to the continuum of two-magnon excitations resulting from this dispersion. Recently it was stressed [7,9] that there exists a second well-defined triplet below the two-magnon continuum near momentum $\frac{\pi}{2}$, but as Fig. 2 indicates, the second triplet occurs only very close to higher states, even for the relatively strong dimerization of $\delta = 0.2$. Therefore it was stated that an alternating chain will not explain the second triplet excitation observed in $(\text{VO})_2\text{P}_2\text{O}_7$ at *all* q-momenta. However, it is known [7] that including frustration, i.e. an antiferromagnetic next nearest neighbor interaction α between \mathbf{S}_i and \mathbf{S}_{i+2} , into the alternating chain model, yields a second well-defined triplet

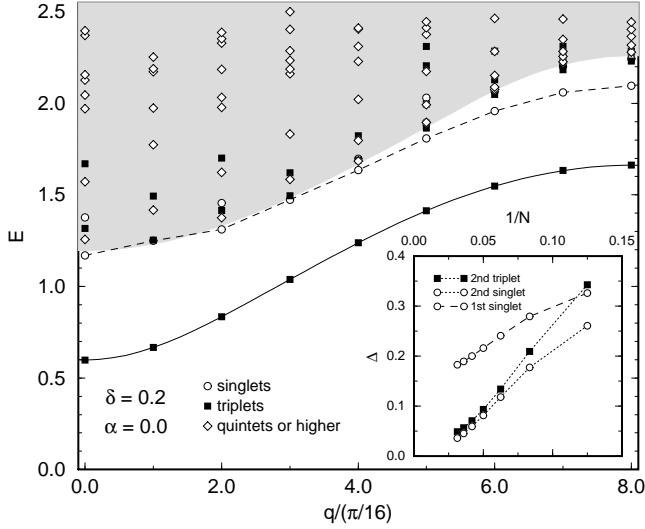


FIG. 2.: Low-energy excitations of the AHC. The inset shows the difference Δ of the 1st and 2nd singlet and the 2nd triplet to the 1st quintet at momentum $\pi/2$ versus inverse chain length.

branch below the continuum in the whole Brillouin zone, provided α is sufficiently strong.

Since an intra-chain frustration is not plausible in view on the structure of $(\text{VO})_2\text{P}_2\text{O}_7$, we will consider a perpendicular coupling of two alternating chains instead:

$$H_{CC} = J_b \sum_{j=1,2} (1 + \delta(-1)^i) \mathbf{S}_{i,j} \cdot \mathbf{S}_{i+1,j} + J_a \sum_i \mathbf{S}_{i,1} \cdot \mathbf{S}_{i,2}. \quad (2)$$

Here j is numbering the chains. As was already suggested in [6], such a coupling in a-direction should be ferromagnetic to explain the observed dispersion. For illustration, in Fig. 3 we plotted a few low-lying energies of a 2×12 system with $\delta = 0.2$ and $J_a = -0.1$. Similar results were obtained for a 3×8 system.

Again we have a well-defined magnon branch. As a guide to the eye we shaded the region where one would expect a two-magnon continuum, approximated here by adding the gap at zero momentum to the magnon dispersion. Close to the continuum edge there are several states: singlets, triplets, as well as quintets. To gain further insight one has to perform a finite-size analysis. For extrapolation to the infinite system we use the following formulas for the lowest singlets and triplets [7,10]:

$$E^S(L) = E^S(\infty) + \left(\frac{B}{L} + C\right)e^{-L/A} \quad (3)$$

$$E^T(L) = E^T(\infty) + \frac{B}{L}e^{-L/A} \quad (4)$$

In Fig. 4 a few low-lying excitations at momenta $(q_b, q_a) = (0, 0)$, $(0, \pi)$ and $(\pi/2, 0)$ are given subject

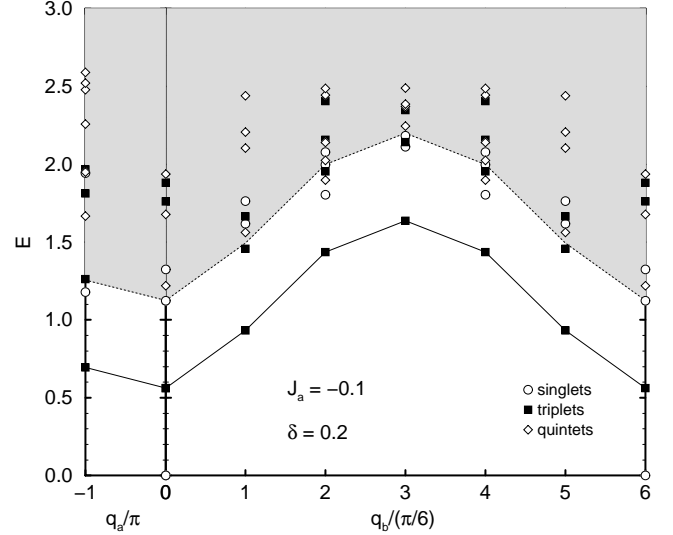


FIG. 3.: Low-energy excitations of two alternating chains coupled ferromagnetically; the parameters are: chain-length = 12, $\delta = 0.2$, $J_a = -0.1$

to the ferromagnetic interchain coupling J_a . At $(0, 0)$ and $(0, \pi)$ the data is extrapolated to infinite system size, while at $(\pi/2, 0)$ results for a 2×16 system are shown, because we have just four different system sizes ($2 \times 4, 8, 12, 16$) at this momentum, making finite-size scaling questionable. Nevertheless, in the inset we tried to extrapolate the second triplet at $(\pi/2, 0)$ to the infinite system, using the ansatz of Eq. (4). The plot indicates that this triplet shows a weak nonmonotonic behaviour, in contrast to the single chain case.

Obviously at momenta $(0, 0)$ and $(0, \pi)$ there are no second triplets below the two-magnon continuum. Just at $(q_b, q_a) = (\pi/2, 0)$ a second triplet stays very close to the continuum edge, and the well-defined singlet excitation, known from the single alternating chain seems to disappear with increasing interchain coupling J_a .

From the above results we conclude that an interchain coupling of this simple type does not qualitatively change the structure of the low-energy excitations compared to the single alternating chain. Excitations are just shifted (as it seems linearly with J_a in most cases), but no new features appear. This is, why we propose another model for $(\text{VO})_2\text{P}_2\text{O}_7$.

We mentioned above that frustration in the alternating chain can lead to a well-defined triplet below the two-magnon continuum. Thus going to the second dimension we include an additional, frustrating coupling J_\times . Then our model Hamiltonian reads

$$H_\times = J_b \sum_{i,j} (1 + \delta(-1)^i) \mathbf{S}_{i,j} \cdot \mathbf{S}_{i+1,j} + J_a \sum_{i,j} \mathbf{S}_{i,j} \cdot \mathbf{S}_{i,j+1} \quad (5)$$

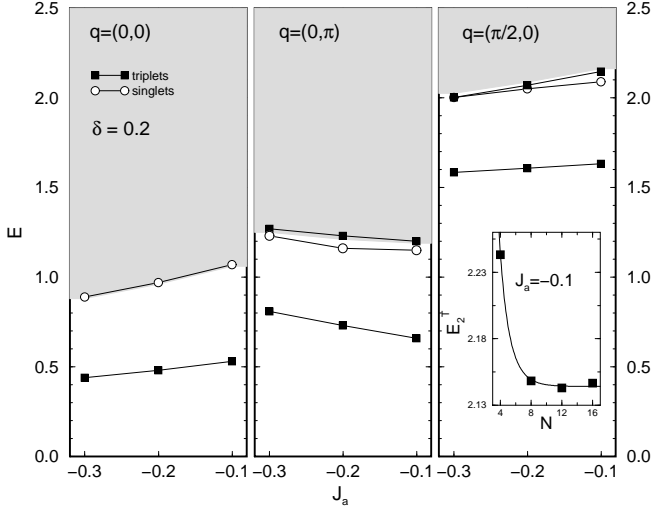


FIG. 4.: Energy of the lowest singlet (open circles) and triplet (filled squares) excitations for different interchain coupling J_a and fixed dimerization $\delta = 0.2$.

$$+J_{\times} \sum_{i,j} (\mathbf{S}_{2i,j} \cdot \mathbf{S}_{2i+1,j+1} + \mathbf{S}_{2i+1,j} \cdot \mathbf{S}_{2i,j+1})$$

(cf. also Fig. 1, lower panel). As yet there is no data available about the strength of such a coupling, but as a first step it seems not unreasonable in view of the oxygen-mediated superexchange paths in $(\text{VO})_2\text{P}_2\text{O}_7$. We assume all exchange integrals to be antiferromagnetic, but still the parameter space is very large. It appears that J_{\times} has to be bigger than J_a to get a ferromagnetic magnon dispersion in a-direction, what is plausible. On the other hand, both couplings should not differ too much for a second triplet branch to exist in the whole momentum space, and should have a sufficient strength. The size of the gap to the first triplet branch is (still) mainly controlled by the dimerization δ .

A good choice of parameters is $\delta = 0.3$, $J_a = 0.4$ and $J_{\times} = 0.425$, for which we diagonalized systems of two, three and four chains with a total number of up to 32 spins and periodic boundary conditions. The low-energy excitations of the 4×8 system are shown in Fig. 5.

Beside two triplet branches we observe also a well-defined singlet, and there might even be a second singlet near momentum $(q_b, q_a) = (\pi/2, 0)$. As the difference between J_{\times} and J_a is small, the dispersion of the triplets is weak in a-direction, in accordance with experiments. We stress that the picture remains qualitatively unchanged going from the 3×8 to the 4×8 system, just the second triplet shifts downwards at momentum $(0, 0)$ with increasing system size. Thus we believe that these features will survive in the infinite system.

To provide some more information on the excitation spectrum, we calculated the dynamical spin structure factor and the integrated spectral weight

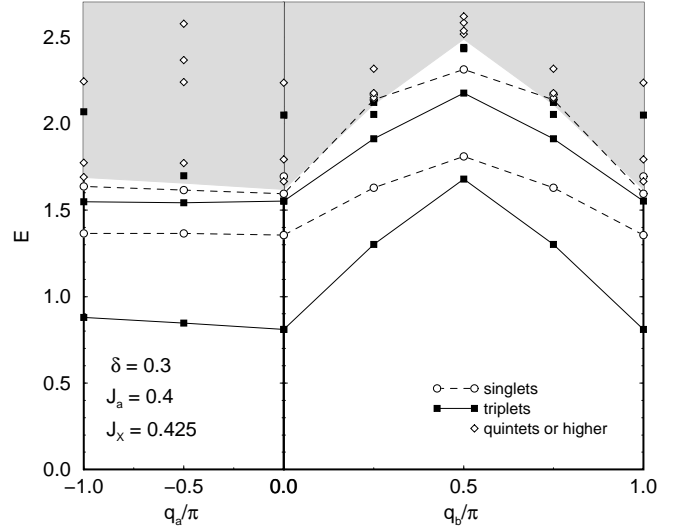


FIG. 5.: Low-energy excitations of the 2D model; the system size is 4×8 , $\delta = 0.3$, $J_a = 0.4$ and $J_{\times} = 0.425$

$$S(\mathbf{q}, \omega) = \sum_n |\langle n | \mathbf{S}^z(\mathbf{q}) | 0 \rangle|^2 \delta(E_n - E_0 - \omega), \quad (6)$$

$$N(\mathbf{q}, \omega) = \int_0^\omega d\omega' S(\mathbf{q}, \omega'), \quad (7)$$

where $\mathbf{S}^z(\mathbf{q}) = \sum_{i,j} e^{i\mathbf{q} \cdot \mathbf{r}_{i,j}} S_{i,j}^z$. In the plot the integral is normalized to one; its real value $N(\mathbf{q}) = N(\mathbf{q}, \infty)$ is noted in each panel.

It seems that we need a finite momentum component in b-direction for the first triplet to have some weight, while the second triplet appears only in a-direction. For comparison take the dashed and solid lines in the upper two panels of Fig. 6, corresponding to momenta (π, x) and $(0, x)$, respectively, that are equivalent in energy.

To summarize, using exact diagonalization methods we have shown that a simple ferromagnetic coupling of alternating Heisenberg chains does not provide two well-defined triplet branches as were observed in inelastic neutron scattering experiments on vanadyl pyrophosphate $(\text{VO})_2\text{P}_2\text{O}_7$. From our experience with frustrated alternating Heisenberg chains, we proposed an alternative model to describe the low-energy physics of $(\text{VO})_2\text{P}_2\text{O}_7$, introducing a frustrating interchain coupling. Due to the large parameter space and the computational effort for sufficiently extended 2D systems, we made no attempt to fix the parameters for $(\text{VO})_2\text{P}_2\text{O}_7$, but showed that the proposed model can describe the general feature of two triplet branches below a continuum of states. These triplets exhibit a ferromagnetic (antiferromagnetic) dispersion in a- (b-) direction. Thus we believe that our model is a good starting point for further analysis.

We thank E. Müller-Hartmann, J. Schliemann, S. Sil and G. Wellein for stimulating discussion. G.B. acknowledges the hospitality at the Universität Bayreuth,

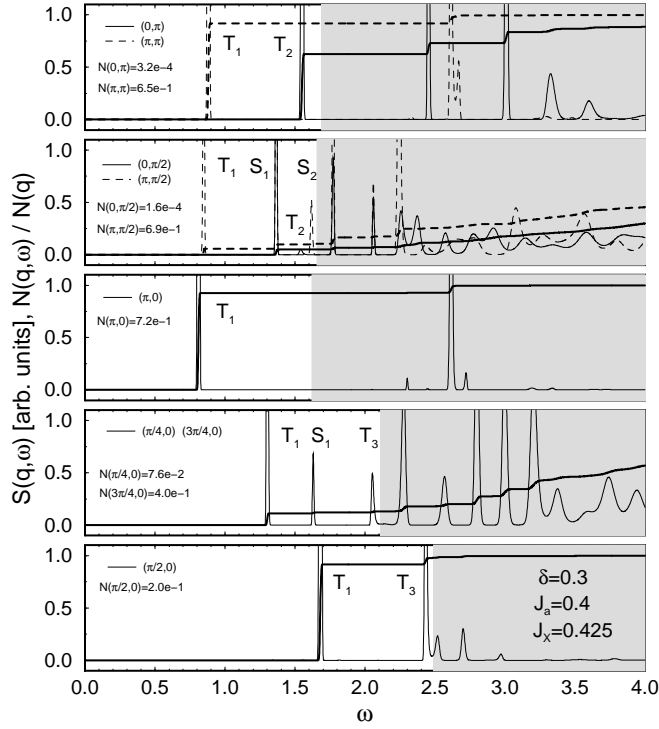


FIG. 6.: Dynamical spin structure factor for the 2D model (4×8 system, $\delta = 0.3$, $J_a = 0.4$, $J_x = 0.425$); the lowest singlet and triplet excitations are classified.

granted by the Graduiertenkolleg 'Nichtlineare Spektroskopie und Dynamik'. Calculations were done on the T3E systems at HLRZ Jülich and HLRS Stuttgart and on the SP2 systems at GMD Bonn and LRZ München.

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